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Graphical Abstract



Abstract

The 'Black Summer' wildfires that affected Australia over the 2019-2020 summer have led to concern over the health effects of exposure to wildfire emissions, and generated a need for means to reduce exposure. Recently, active green infrastructure has been implemented in cities to assist in the removal of urban air pollution, however the filtration of wildfire emissions has not been previously tested. Here, we field trial botanical biofiltration for the reduction of elevated air pollutant concentrations associated with Black Summer. Two active green walls were installed in outdoor environments in Sydney over Black Summer, with the concentrations of NO₂, O₃ and PM_{2.5} in ambient and filtered air streams monitored over 14 days with elevated air pollution concentrations due to wildfire emissions. Average pollutant single pass removal efficiencies of 63.17%, 38.79% and 24.84% for NO₂, O₃ and $PM_{2.5}$ respectively were recorded, with clean air delivery rates of 558.90 m³/h, 343.19 m^{3}/h and 219.77 m^{3}/h for NO₂, O₃ and PM_{2.5} respectively for each 5 m^{2} biofilter wall. Weak negative associations were observed between the removal efficiency of NO₂ and PM_{2.5} and their corresponding ambient concentrations. Strategic employment of botanical biofiltration may thus be of value in reducing wildfire emissions in sensitive populations.

Keywords: botanical biofilter; green wall; living wall; green infrastructure; bushfire

Highlights:

- Active green walls demonstrated removal of NO₂, O₃ and PM_{2.5} from wildfire smoke.
- NO₂ was removed most efficiently, with a single pass removal efficiency of 63.17%.
- Clean air delivery rates of 220-559 m^3/h were achieved for all pollutants.
- NO_2 and $PM_{2.5}$ removal rates were negatively correlated with ambient concentrations.

1. Introduction

Over recent decades, fire weather seasons have increased in frequency and intensity across a quarter of the earth's vegetated surface, and the global burnable area affected by long season fire has doubled (Jolly et al., 2015). This trend will likely be associated with an increase in population exposure to wildfire smoke and corresponding health impacts. Strong correlations have been identified between wildfire smoke exposure and both respiratory morbidity and all-cause mortality, and have been attributed to an estimated 339,000 annual deaths worldwide (Analitis et al. 2012; Johnston et al. 2012; Stauffer et al. 2020). These effects are likely to be more pronounced in wildfire 'hot spots' such as Australia, wherein recent climatic shifts have been associated with considerably increased wildfire frequencies, particularly during dry summers (Dutta et al. 2016).

Wildfire smoke contains a mixture of harmful pollutants. Fine particulate matter (PM_{2.5}: particles with an aerodynamic diameter less than 2.5 μ m) is a major constituent of wildfire emissions (Vicente et al. 2013), and has well-known harmful properties (Xing et al. 2016). Additionally, wildfire smoke contains nitrogen dioxide (NO₂; Mebust et al. 2011) and ozone (O₃; Jaffe and Wigder 2012) which are both strong oxidants associated with respiratory symptoms and diseases (Latza et al. 2009; Nuvolone et al. 2018)

The *Black Summer* wildfire natural disaster that affected Australia over the summer between December 2019 and February 2020 (Borchers Arriagada et al. 2020; Vardoulakis et al. 2020; Walter et al. 2020; Yu et al. 2020) burned an estimated 17 million hectares of land across New South Wales, Victoria, Queensland, Western Australia, South Australia and the Australian Capital Territory (Parliament of Australia 2020), with smoke plumes extending as far New Zealand (NASA 2020). The scale and duration of population exposure to wildfire emissions were unprecedented (Walter et al. 2020), and have been associated with 417 deaths, 1124 hospital admissions for cardiovascular symptoms, 2027 hospital admissions for respiratory symptoms and 1305 emergency department attendances for asthma symptoms (Borchers Arriagada et al. 2020). With the predicted increases in severity and frequency of wildfires, and increasing awareness of the adverse health effects of

wildfire smoke, questions are now being raised on what effective actions can be taken to reduce exposure.

Over the past two decades, a range of active botanical biofiltration systems has been developed to mitigate ambient air pollution (see Irga et al. 2020 for descriptions of several systems). These systems use mechanically generated, active airflow usually supplied by low power fans — to pass a contaminated airstream through a vertical plant growth substrate and foliage (Pettit et al 2018a). As the air stream passes through the system, a range of air pollutants can be removed through physical, biological and chemical mechanisms; particulate matter (PM) is filtered by the substrate and root system (Pettit et al. 2017) and gaseous pollutants, such as volatile organic compounds (VOCs; Pettit et al. 2019a), NO₂, and O₃ (Pettit et al 2019b) adhere to specialised substrate adsorbents (Pettit et al 2018b) or become degraded by the plants' root zone microbial community (Pettit et al 2018a). Previous work has demonstrated the capacity of active botanical biofilters to effectively remove pollutants from indoor environments (Wang and Zhang 2011; Pettit et al. 2019c). Consequently, a range of commercial active green infrastructure designs have been implemented in urban settings to assist in the removal of urban air pollutants (Irga et al. 2020). Previous work has focused on the biofiltration of anthropogenic pollutants; however there has been no assessment of the biofiltration of wildfire-associated air pollutants in open, outdoor environments. The Black Summer wildfires provided a unique opportunity to assess the biofiltration of wildfire emissions by such systems. The air pollution from *Black Summer* has been compared to that of the most polluted mega cities (Vardoulakis et al. 2020), consequently research that contributes towards the understanding of how these systems perform in highly polluted environments is of considerable value.

The aim of the current research was thus to assess the capacity of an active green wall biofilter to filter NO_2 , O_3 and $PM_{2.5}$ from wildfire polluted ambient air, and thus to provide 'clean air' during wildfire events, and secondly, to examine the effect of ambient pollutant concentration on filtration efficiency during wildfire events.

2. Methods

2.1 Active green wall description

The active green walls (Figure 1) used in this experiment were comprised of five plenums (1 x 1 m, depth 0.15 m; Figure 1), providing a front face surface area of 5 m². Each plenum had four openings on its front face (63.6 cm² cross sectional area), that were each connected to a modular botanical biofilter (Breathing Wall; Junglefy P/L, Sydney Australia). Biofilter modules were made from recycled low-density polyethylene containing a coconut husk-based growth substrate. There were 16 holes on the front (polluted air inlet) face of these modules into which plants of the following species were grown: Westringia fruticosa (coastal rosemary), Myoporum parvifolium (dwarf native myrtle), Stobilanthes anisophyllus (goldfussia) and Nandina domestica (heavenly bamboo). These species were selected for their survivability under normal Sydney environmental conditions. Internal linings of highdensity polyethylene shade cloth retained the roots of the plants and the plant growth substrate within the modules. The outlet face of the modules had central ports that connected them to the plenums. Air was driven through the plant foliage, growth substrate and the plenum by two fans per plenum (NF-F12, Noctua, Austria; volumetric flow rate 186.70 m³/h; internal diameter 120 mm; power consumption

4.32 W) located on the rear face of each plenum. These fans were chosen as they provided a volumetric flow rate similar to Irga et al.'s (2017) optimised PM_{2.5} SPRE flow rate when systems were standardised by filter area. Fans were operated from 6:00 am to 6:00 pm with a period without fan operation overnight providing temporal independence for daily samples. Two identical active green wall systems were used, located near a roadside (Hills Motorway) in Sydney, Australia. The walls were separated by >50 m to provide spatial independence. As these green walls were located outdoors, they were exposed to ambient air pollutant concentrations comprised of both vehicular exhaust and wildfire smoke.





Figure 1. a = One of the active green walls used in this study; b and c = Plenum used to hold the active green wall modules and isolate the effluent airflow. b) shows the front (polluted air inlet) face of the plenum without planted modules attached. c)

shows the rear (filtered air outlet) face. Fans were housed within the air outlets to produce active airflow. Five plenums were placed side-by side horizontally to create 5 m^2 active green walls.

2.2 Wildfire events

As traffic emissions are normally the major contributors to NO_x and PM_{2.5} pollution in Sydney (Cowie et al. 2019; Crawford et al. 2017; Paton-Walsh et al. 2019), diurnal variability in ambient pollution concentrations was influenced by temporal fluctuations in traffic volume independent of wildfire emissions. Variation in traffic thus represented a cyclic pattern in the ambient pollutant concentration, whereby contributions from wildfire emissions followed a random pattern, predominantly influenced by fire and wind characteristics. A time series analysis was thus conducted using the ambient $PM_{2.5}$ concentration recorded at each wall (see 2.3 Sampling regime) as a surrogate variable for general air pollution, as it is strongly associated with both traffic and wildfire emissions (Forehead et al. 2020). The 14 days where the random residual variation in PM2.5 concentration exceeded the maximum cyclical variation were thus deemed to be 'wildfire days' (Figure 2). To ensure the emissions detected on these days were primarily sourced from wildfires, the data was cross-checked against the average 24 h PM_{2.5} concentration collected from the New South Wales Government Department of Planning Industry and Environment (see Supplementary Material) and historical air quality data (Johnston et al. 2011). The average PM_{2.5} 24 h concentrations across the NSW DPIE's Sydney air quality-monitoring network on these days were all within the 99th percentile of historical data from 1994-2007. Over this 13 y period, all days with PM_{2.5} concentrations at this level were attributable to landscape fire (hazard reduction burns and wildfire) smoke, with a single exception caused by a dust storm (Johnston et al. 2011). Additionally, the wildfire days identified in Black Summer were also crosschecked with announcements from the NSW Rural Fire Service, which all reported fires in close proximity to Sydney on these days.



Figure 2. The observed and decomposed time series of the $PM_{2.5}$ concentration ($\mu g/m^3$). The grey line across the 'random' variation represents the maximum cyclical variation in $PM_{2.5}$ concentrations. Wildfire days were readily identified from the 'random' trend.

2.3 Sampling regime

The concentrations of NO₂, O₃ and PM_{2.5} were detected by a network of Aeroqual AQY1 micro air quality monitoring systems (Aeroqual, Auckland, New Zealand). One AQY1 system was placed on the end of each active green wall and these measured the ambient pollutant concentrations throughout the experiment, and provided a spatially-relevant baseline against which to compare filtered air pollutant concentrations. AQY1 units were also placed inside each of the five plenums within each green wall. As the plenums isolated the filtered effluent airstream, these instruments recorded pollutant concentrations in the filtered effluent air streams from each independent plenum. Each instrument logged the average concentrations of NO₂, O₃ and PM_{2.5} every five minutes across the entire *Black Summer* experimental period (December 2019-February 2020).

The airflow through each of the plenums was quantified with a VelociCalc Air Velocity Meter 9545 (TSI Incorporated; Shoreview, Minnesota, USA). The air velocity through each of the effluent vents which was multiplied by the cross sectional area of the vent openings to calculate the volumetric flow rate through each of the plenums.

2.4 Data analysis

Pollutant single pass removal efficiencies (SPREs) were calculated by comparing the ambient pollutant concentrations to those in the isolated effluent air stream from time-matched samples. SPREs were taken as a function of the volumetric flow rate to estimate the clean air delivery rates (CADRs) of each pollutant provided by the 5 m² active green walls (CADR = SPRE x biofilter airflow rate).

To assess the monotonicity of the relationship between SPREs and ambient concentrations, Spearman's correlations were conducted for each pollutant.

3. Results

Pollutant concentrations in the ambient and filtered airstreams for wildfire days are shown in Figures 3a-c. In all cases, lower pollutant concentrations were observed in the effluent air streams than in ambient air, however the magnitude of these differences were not consistent across pollutants. Ambient concentrations of all pollutants varied widely, and frequent, dramatic changes in concentrations occurred, likely a consequence of the meteorological influences on smoke transportation from the wildfires. Across the whole sampling period, maximum five-minute-average-concentrations of 178.6 ppb, 59.4 ppb, and 774.7 μ g/m³ were detected for NO₂, O₃ and PM_{2.5} respectively. Given Sydney's normally good air quality, these values were extraordinary.



Figure 3a. The concentrations of NO_2 , O_3 and $PM_{2.5}$ on days with elevated pollutant concentrations due to wildfire emissions. Average concentrations are shown for the ambient pollutant concentrations and the concentrations in the filtered effluent airstream.



Figure 3b. The concentrations of NO_2 , O_3 and $PM_{2.5}$ on days with elevated pollutant concentrations due to wildfire emissions. Average concentrations are shown for the ambient pollutant concentrations and the concentrations in the filtered effluent airstream.



Figure 3c. The concentrations of NO_2 , O_3 and $PM_{2.5}$ on days with elevated pollutant concentrations due to wildfire emissions. Average concentrations are shown for the ambient pollutant concentrations and the concentrations in the filtered effluent airstream.

As the focus of this work was to assess the removal of wildfire emissions, lower limit thresholds were assigned to each pollutant to exclude data from the analysis that was unrepresentative of elevated pollutant concentrations associated with wildfires. For NO₂ and PM_{2.5}, data where the ambient concentrations were less than 19 ppb and 25 μ g/m³ respectively were excluded. These values represent the World Health Organisation's recommended annual NO₂ exposure and 24-hour PM_{2.5} exposure limits (World Health Organisation 2018). As Sydney usually experiences ozone concentrations much lower than the WHO recommendations (Paton-Walsh et al. 2019), a lower limit threshold of Sydney's annual mean O₃ concentration of 18.5 ppb was used (Paton-Walsh et al. 2019). All subsequent analyses used only data where ambient pollutant concentrations were greater than these corresponding thresholds.

 NO_2 was removed most efficiently by the active biofilters, with an average SPRE of 63.17%, while O_3 and $PM_{2.5}$ were removed with lower removal efficiencies of 38.79% and 24.84% respectively. These were converted to clean air delivery rates by multiplying the SPRE by the volumetric flow rate, which was 884.8 m³/h through each 5 m² active green wall, producing average CADRs of 558.9 m³/h for NO_2 , 343.2 m³/h for O_3 , and 219.8 m³/h for $PM_{2.5}$.

Spearman's rank correlations were used to assess the monotonicity of the relationships between the ambient concentrations and the SPREs of each pollutant (Figure 4). Although we detected no association for O₃, NO₂ demonstrated a weak negative association (p = < 0.01, $\rho = -0.158$, n = 919) as did PM_{2.5} (p = < 0.01, $\rho = -0.251$, n = 1075).

Figure 4. The ambient concentration of pollution against the corresponding SPRE of a $= NO_2$; $b = O_3$; $c = PM_{2.5}$.

4. Discussion

This work represents the first trial of an outdoor, infrastructure scale filtration system of any type to ameliorate high concentrations of wildfire-associated air pollutants. The pollutants were removed with different removal efficiencies, and whilst the mechanisms of pollutant removal still require further research, it is probable that the contribution of each removal mechanism varies with the chemical and physical properties of each pollutant, as well as the biotic and abiotic components of the biofilter system (Pettit et al. 2019a). Of the three pollutants assessed, NO₂ was removed most efficiently, with an average SPRE of 63.17%. Although the SPRE of NO₂ was more variable at lower ambient concentrations, whereby small differences in the NO₂ concentration in the influent or effluent had a disproportionately large impact on the SPRE, the removal efficiency was relatively consistent throughout the range of observed ambient concentrations. Comparatively, O₃ was removed less efficiently, at an average SPRE of 38.79%. Although the botanical biofiltration of NO₂ and O₃ has been observed in laboratory studies using spiked concentrations of pollutants (Pettit et al. 2019b), this work represents the first observation whereby a constant removal of wildfire associated pollutants from a naturally generated influent air stream by active green wall biofilters has been demonstrated. While gaseous pollutants are removed by several processes, usually beginning with dissolution in to the aqueous phase of the filtration matrix, the fate of the filtered pollutants, and their ramifications for the active green wall system, remains unclear. Previous work has noted the potential production of nitric acid within the growth substrate as NO₂ combines with irrigation water (Zheng et al. 2016). The simultaneous biofiltration of O₃ and NO₂ may have enhanced pH control due to the generation of alkaline products from O₃ biofiltration (Maldonado-Diaz and Arriaga 2015). Although it was not our intention to assess filtration products in this study, any changes in substrate pH were insufficient to visibly affect plant health or influence system performance.

When standardised by substrate volume, the NO₂ and O₃ CADRs observed in this study were substantially higher than those observed in the laboratory (Pettit et al. 2019b), most likely due to the use of different ventilation systems and pollutant inlet concentrations between the studies, with the current measurements likely providing a more accurate estimate of the *in situ* air cleaning potential of the green wall biofilters.

The PM_{2.5} SPRE was lower than that reported in laboratory studies. Irga et al. (2017) reported a PM_{2.5} SPRE of 48% for diesel smoke for an equivalent active green wall biofiltration system. The composition of the tested PM_{2.5}, including both the size-distribution of particles and chemical composition, is likely to have caused these differences. Pettit et al. (2017) found that larger particles, 1–2.5 µm in diameter, were filtered much more efficiently by green wall biofilters than smaller particles in the 0.3-1 µm diameter class. The instruments used in the current study did not facilitate the discrimination of particles smaller than PM_{2.5}, however, detection of smaller size fractions and understanding the ambient size distribution within the PM_{2.5} particle size class in wildfire emissions would allow for a more critical assessment of particulate filtration. Although less efficient than laboratory estimates, the PM CADRs observed in this study are nevertheless of value, as they demonstrate the practical removal of particle compositions from a real urban environment. PM_{2.5} biofiltration by active green walls has been demonstrated in indoor environments (Pettit et al. 2019), however such work has been limited to the short-term (< 1 h) removal of spiked or ambient PM. The current trials build on this work, and demonstrate that successful, prolonged biofiltration is possible. Although the PM_{2.5} SPRE reported in this study

were less efficient than laboratory estimates achieved by a similar active green wall system and a Minimum Efficiency Reporting Value (MERV) 11 pleated panel HVAC (heating ventilation and air conditioning) filter (Irga et al. 2017), the PM_{2.5} SPRE observed in this study is highly valuable as it reflects the *in situ* treatment of a unique source of PM. As seen in heavy vehicle diesel particulate filters, the impact of ash and soot accumulation in filters, both independently and as a mixture, potentially compromises the filters efficiency (Kimura et al. 2006). When soot pollutants accumulate in filters, these air pollutants can potentially be regenerated.

Previous work has revealed that PM is filtered by the matrix created by the plant growth substrate and plant root system (Pettit et al. 2017), however the effects of long term PM filtration, particularly at high concentrations, are yet to be tested. The dynamic nature of the system, including irrigation regimes and plant and microbial activity, make it difficult to estimate the possible effects on biofilter airflow rates that may results from long-term PM accumulation within the matrix.

The CADRs of all pollutants indicate that even relatively small active green wall systems can provide considerable volumes of filtered air, which could provide realistic improvements in environmental quality if such systems are strategically deployed, and the filtered airstream is managed in such a manner so as to delay atmospheric dilution.

Although the removal efficiencies of NO₂ and PM_{2.5} were negatively associated with their respective ambient concentrations, the strength of these associations was weak in both cases, and is it likely that other variables that were not measured in this study may have stronger associations with removal efficiency. The moisture level of the plant growth substrate is an aspect that is associated with removal rates of some pollutants (Abdo et al. 2019; Pettit et al. 2019a), and as these active green walls were exposed to rain and subject to a particular irrigation regime, it is likely that the substrate moisture level varied throughout the trial period, and may have influenced filtration efficiency. Other temporal effects including plant growth and PM accumulation over time may have altered the substrate matrix and be linked with variability in pollutant removal efficiency, however no long term *in situ* studies of the simultaneous botanical biofiltration of NO₂, O₃ and PM_{2.5} are available. As this experiment focused on the removal of wildfire emissions under ambient in situ conditions, ambient temperature and humidity were not manipulated throughout the experiment. It is possible that variations in temperature and humidity may have lead to variations in SPREs throughout the trial, and these effects would be worth including in future laboratory studies. While pollutant removal remains a complicated process influenced by a range of physical, chemical and biological properties and processes, it is possible that at higher concentrations there is a saturation effect whereby the removal efficiency is reduced. Nonetheless, the ambient pollutant concentration remains an important consideration of system performance, and should be explored in conjunction with temporal effects in future research. In some cases in the current trials, pollutant concentrations in the effluent air stream were greater than in the ambient air stream, however in all cases where this occurred, the ambient concentrations of pollutants were generally very low and absolute differences in pollutant concentrations between the filtered and ambient air were comparatively small.

In extreme air pollution events such as *Black Summer*, any air pollution mitigation strategies are clearly of value. The current findings suggest that active green walls in targeted locations may be a valuable adjunct to the wearing of facemasks and staying indoors. The use of indoor air cleaners during wildfire events

has been identified as an effective means by which to reduce exposure to emissions (Barn et al. 2016), and the use of active green walls in indoor settings may offer a similar effect. Although this trial occurred in an outdoor setting, wherein the rate of pollution removal was small compared to the rate of emissions, the use of active green walls in indoor settings may be a more appropriate means of providing enhanced air quality and reducing occupant exposure to wildfire emissions. These strategies may be particularly useful in indoor environments that remain susceptible to wildfire smoke infiltration, as the protection provided by a building is dependent upon building construction and the degree of infiltration of outdoor air (Barn et al. 2016).

While the air pollution profile generated by wildfire emissions differ greatly from those that are anthropogenically-derived, the gross pollution concentrations observed in this study are comparable to ambient environments in highly polluted mega cities (Vardoulakis et al. 2020), and thus the removal rates reported in this study may also be achievable in these locations. The critical need for air quality improvements in some highly polluted urban areas has led to the development of several novel air purifying towers (i.e. see Cao et al. 2014 and Smisek 2018) and this has highlighted the potential of up-scaled systems to impact the surrounding ambient air quality. Upscaling active green walls in hotspot locations within large, polluted cities and assessing their influence on the ambient air quality (in addition to their other environmental benefits, see Perini and Rosasco 2013) is a valuable area of future research. The low cost of these systems compared to conventional air filtration devices, along with their 'green credentials' favours the scalability of these systems. Increasing the size of active green walls in open urban contexts would not only proportionately increase the CADR provided by each wall, but may, in some cases, create a barrier between the emission source and the relevant receiver on the leeward side of the wall (Abhijith et al. 2017). No study to date has measured the combined synergistic impact of the barrier effect and CADR on the surrounding ambient air quality and this remains an important consideration in subsequent studies.

5. Conclusion

Between December 2019 and February 2020, Australia experienced elevated air pollution due to extensive emissions from the *Black Summer* wildfires. The research presented here demonstrated that a green wall biofilter with active air flow drawing untreated air through the plant foliage and growth substrate was able to filter NO_2 , O_3 and $PM_{2.5}$ during periods of high pollution levels associated with wildfire emissions. Across the observed pollutant concentrations, NO_2 was removed with greater efficiency than O_3 and $PM_{2.5}$. As these pollutant concentrations are comparable with those in mega cities with poor air quality, future work should trial such systems in these environments and assess the impacts of filtration on the ambient air quality.

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Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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